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(54) STORABLE COMPOSITIONS, FOR DILUTION WITH WATER TO FORM ANTI-MICROBIAL SOLUTIONS

(71) We, SCHULKE & MAYR GMBH, a Company organized under the Laws of the Federal Republic of Germany, of Robert-Koch-Strasse 2, 2000 Norderstedt, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention concerns storable compositions, for dilution with water to form anti-

microbial solutions.

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Organic peracids are known to display outstanding anti-microbial activity, but their use as anti-microbial agents gives rise to certain problems in actual practice, broadly speaking because they tend to be inadequately stable and/or inadequately water-soluble. Thus, certain percarboxylic acids, such for example as perbenzoic acid, are unstable even in solid form, and no satisfactory way has therefore been found of putting them upor the market. There are other percarboxylic. acids which are stable, such as para-tertbutyl-perbenzoic acid and para-methoxyperbenzoic acid, but these are soluble in water only with difficulty. Consequently, even if it be possible ultimately to form an aqueous solution containing an anti-microbiallyeffective concentration of such difficultlysoluble peracids, it is in practice really rather difficult to establish such a concentration because of the low rate at which these peracids dissolve.

There are indeed certain lower aliphatic mono-percarboxylic acids, such as peracetic and perproponic acid, which are capable of forming stable aqueous solutions, provided that the peracid content of these solutions is high. However, although stable, such concentrated peracid solutions possess certain disadvantages—firstly because they have an extremely pungent odour, which makes them unpleasant, difficult and even hazardous to

use; and secondly because when spilled these concentrated solutions cause corrosion or other damage to materials, and/or violent decomposition reactions. These problems might be mitigated by using more dilute solutions of such lower aliphatic mono-percarboxylic acids, but unfortunately such dilute solutions even though they have a satisfactory anti-microbial disinfectant activity are not stable over any prolonged period, and for that reason cannot be marketed as such.

It might be thought that another way of mitigating the drawbacks of the various organic peracids would be to form neutral or alkaline salts of them, but it has been found that generally-speaking organic peracids are not stable in the neutral and alkaline ranges over any reasonably prolonged period of time, so again this provides no satisfactory practical solution to the problem of using them for their anti-microbial activity.

There is therefore an obvious need for some kind of solid formulation which is adequately stable during manufacture, storage and marketing, yet which when dissolved in water immediately before use will quickly yield an aqueous solution with a reasonably high concentration of percarboxylic acid and thus a good anti-microbial disinfectant activity.

Since it is commonplace for many cleaning purposes to use alkaline solutions to promote the cleaning action, and clearly it would be desirable to impart an anti-microbial, disinfectant activity to such alkaline cleaning solutions by incorporating organic peracids therein, there is also a need for formulations of organic peracids which display an alkaline reaction and thus will be stable in an alkaline medium, even though the organic peracids themselves are not, as already indicated above.

We have now surprisingly found that it is possible to form a stable, solid mixture comprising both certain acyloxy-aromatic-carboxylic acids (and/or salts thereof) as well 45

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as one or more H₂O₂ source(s), and such compositions when dissolved in water can yield an alkaline-reacting solution which displays outstanding anti-microbial, disinfectant activity, and also at least some degree of antifungal activity—this being particularly sur-prising, since organic peracids in general do not usually possess any significant activity against fungi.

According to one aspect of the invention, therefore, there are provided storable compositions, for dilution with water to form (A) at least one acyloxy-aromatic-carboxylic acid of the general formula:

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[in which the carboxy group is in the ortho-, meta- or para-position relative to the acyloxy radical; and either R represents a lower alkyl or lower oxa-alkyl radical, optionally substituted by a hydroxy group, a lower alkoxy group, a lower acyloxy group, a carboxy group or a halogen atom, or R represents a phenyl group, optionally substituted by one or more hydroxy group(s), halogen atom(s), lower alkyl group(s) and/or lower alkoxy group(s)] and/or at least one water-soluble alkali metal, alkaline earth metal or ammonium or alkylsubstituted (including quaternary) ammonium salt(s) thereof; and

(B) one or more H₂O₂ source(8). The components of this solid mixture, namely the acyloxy-aromatic-carboxylic acids of general formula I above and/or salts thereof forming component (A) and the H₂O₂ source(s) forming component (B) thereof, do not react together, to any significant extent, while in the solid state within the mixture during manufacture, storage and marketing-but they are sufficiently reactive so that upon dissolving the mixture in water or similar aqueous systems they will immediately commence to react with each other, thus forming a solution containing both percarboxylic acid and hydroxy-aromatic-carboxylic acid. This resultant solution displays a highlyeffective, quick-acting anti-microbial, disin-fectant activity derived from the percarboxylic acid, combined with a long-term anti-microbial activity derived from the hydroxy-aromaticcarboxylic acid.

While we do not wish to be limited by any theoretical considerations, it appears that when the solid composition is dissolved in water, a reaction commences between the acyloxy-aromatic-carboxylic acid and the hydrogen peroxide liberated from the H₂O₂

source, which is thought to continue for some time even after a clear solution has been formed, and this reaction cleaves the ester grouping, forming percarboxylic acid on the one hand and free hydroxy-aromaticcarboxylic acid on the other hand. The acyloxy-aromatic-carboxylic acids of general formula I above, and/or their salts, do not however apparently undergo this cleavage very rapidly but only over a certain period of time, and this it is thought is probably fundamental to the success of the compositions of this invention, since the steady but long-drawn cleavage of the ester grouping ensures a high peracid concentration in the aqueous solution over a fairly long period of time, despite the solution being alkaline which otherwise would rule out the maintenance of so high a peracid concentration over a comparable period of

Whether or not that be the correct theoretical explanation, we have found that it is not possible to achieve a comparable, satisfactory aqueous anti-microbial, disinfectant solution by incorporating the reaction products as such in water. Thus if the percarboxylic acid and the hydroxy-aromatic-carboxylic acid are simply put together, rather than being formed in situ, the results obtained are much inferior to those secured by the practice of the present invention. Neither in solid nor in liquid form are such mixtures stable, and particularly not within the pH range of from 4 to 12 which is preferred in the solutions obtained in accordance with the present invention.

The choice of the components (A) and (B) for use in the compositions of this invention depends upon various factors. It may be noted that the acyloxy-aromatic-carboxylic acids of general formula I above (and/or their salts) display an especially high water-solubility in the case of those wherein the radical R is a lower alkyl radical substituted by a carboxy group, especially a free, unsalified carboxy group. It may also be noted that, generallyspeaking, the compositions provided according to this invention will dissolve most quickly if the acyloxy-aromatic-carboxylic acid com- 105 ponent (A) is present in the form of one or more of its water-soluble salts.

It is an empirically-ascertained fact, which moreover is in line with the theoretical explanation tentatively advanced above, that 110 the cleavage of the ester grouping in the acyloxy-aromatic-carboxylic acid of general formula I above (and/or its salts) is promoted in an alkaline medium. Unless indeed it is for some reason desired to delay the hydrolysis of the ester, it is therefore desirable that the aqueous solution formed upon dissolution of the composition of the invention should have a p-value in the range of from 8 to 12. That pH range corresponds in fact to that

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of many of the commonplace alkaline cleaning solutions, to which therefore the compositions of the invention can be added. Even however though the compositions of the invention are to be added to water or other substantially neutral aqueous systems, it may be possible to form an aqueous solution from the composition which has pH in this range of from 8 to 12 by adding sufficient amounts of appropriately-chosen components, thus the more strongly alkaline salts of the acyloxyaromatic-carboxylic acid(s) of general formula I above and/or the more strongly alkaline salts (to be described hereinafter) serving as H₂O₂ source(s).

Equally however the compositions of this invention may also advantageously be incorporated in substantially neutral aqueous solutions, the term "substantially neutral" being used herein to mean a solution with a pH value in the range of from 4 to 8. Indeed, from the viewpoint of the stability of the organic peracids released into the solution, such a substantially neutral pH range is more advantageous than an alkaline pH range. In order to ensure that the composition when added to water or some other substantially neutral aqueous system will result in a solution still within the substantially neutral pH range, it is only necessary to ensure that the proportion of alkaline-reacting components in the composition is such that, after cleavage of the ester, the alkaline ingredients remaining are then consumed in neutralisation of the percarboxylic acid and of the hydroxy-alkylcarboxylic acid, so that consequently a neutral solution is achieved:

The radical R in the acyloxy-carboxylic acids of general formula I above (and/or their salts) used in the compositions of this invention can with particular advantage be a methyl, ethyl, hydroxy-methyl, acetoxymethyl, methoxy-methyl, ethoxy-methyl, 1hydroxy-ethyl, 2-hydroxy-ethyl, 2-carboxyethyl, 3-carboxy-propyl, 3-carboxy-2-oxapropyl, chloro-methyl, phenyl, 2-methyl-phenyl, 3-methyl-phenyl, 4-methyl-phenyl, 2-tert-butyl-phenyl, 3-tert-butyl-phenyl, 2-methoxy-phenyl, tert-butyl-phenyl, methoxy-phenyl, 4-methoxy-phenyl, ethoxy-phenyl, 3-ethoxy-phenyl, 4-ethoxy-phenyl, 2-chloro-phenyl, 3-chloro-phenyl, 4-chloro-phenyl, 2-hydroxy-phenyl, 3-hydroxyphenyl or 4-hydroxy-phenyl group.

Preferred acyloxy-aromatic-carboxylic acids of general formula I above, for use in the compositions of this invention, are for instance, acetyl-, propionyl-, benzoyl-, (4methoxy - benzoyl) - (3 - chlorobenzoyl)-, (4 - tert - butyl - benzoyl)- and (4 - methylbenzoyl) - salicylic acid; 3-acetoxy-, 3propionyloxy-, 3-benzoyloxy-, 3 - (4 - methoxy - benzoyloxy)-, 3 - (3 - chloro - benzoyloxy)-, 3 - (4 - tert - butyl - benzoyloxy)-

and 3 - (4 - methyl - benzoyloxy) - benzoic acid; and 4-acetoxy-, 4-propionyloxy-, 4benzoyloxy-, 4 - (4 - methoxy - benzoyloxy), 4 - (3 - chloro - benzoyloxy)-, 4 - (4 - tertbutyl - benzoyloxy) and 4 - (4 - methylbenzoyloxy) - benzoic acid.

Especially preferred acyloxy - aromaticcarboxylic acids of general formula I are acetyl-, benzoyl-, (4 - methoxy - benzoyl)and (4 - tert - butyl - benzoyl) - salicylic acid; 3-acetoxy-, 3-benzoyloxy- and 3 - (4methoxy - benzoyloxy) - benzoic acid; and 4-acetoxy-, 4-propionyloxy-, 4 - benzoyloxyand 4 - (4 - methoxy - benzoyloxy) - benzoic

When the acyloxy - aromatic - carboxylic acid of general formula I is used in the compositions of the invention as component (A) in the form of one of its salts, suitable watersoluble salts thereof are lithium, sodium, potassium, magnesium and also ammonium, substituted ammonium and quaternary ammonium salts. The substituted (including quaternary) ammonium salts employed will preferably be those in which 1, 2, 3 or even all 4 hydrogen atoms have been replaced by alkyl

The preferred salts for use in the compositions of this invention are the sodium and/or potassium and/or ammonium and substituted (including quaternary) ammonium salts.

The use of such water-soluble salts of the acyloxy-aromatic-carboxylic acids of general formula I above in the compositions of this invention is particularly preferred not only 100 because they are simple to prepare and display both a high solubility and also a high rate of dissolution in water, but also because they have a higher melting point than the acids from which they are derived. A relatively 105 high melting point is a matter of practical significance, since otherwise the solid composition is liable to undergo agglomeration, if for instance it is subjected to solar radiation or any other source of relatively high 110 temperature during storage. The salts however are substantially free from that kind of defect—thus for instance the sodium salt of benzoylsalicylic acid has a melting point greater than 300°C. Thus the use of the salts 115 of the acyloxy-aromatic-carboxylic acids of general formula I in the compositions of this invention ensure problem-free storage of such compositions, without fear of the agglomeration which otherwise might take place, and 120 very disadvantageously impair the rapid and complete dissolution of the compositions according to the invention in water or aqueous systems.

The compositions of the invention must also 125 contain an H2O2 source—that is to say a compound which when the compositions are added to water is capable of generating H₂O₂

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in the formed solution. Suitable H₂O₂ sources include alkali metal and ammonium perborates, percarbonates, perphosphates, peroxides, percarbamides and alkali metal and ammonium salts of persulphuric acid (Caro's acid) and of peroxydisulphuric acid.

The relative proportions in the compositions of this invention between the acyloxy-aromatic-carboxylic acids serving as component (A) and the H₂O₂ source is not highly critical, and in fact can vary within broad ranges. Normally however the composition will contain components (A) and (B) within a molar weight ratio of from 1:10 to 10:1. The preferred molar weight ratio is within the range

of from 1:4 to 4:1.

The composition according to the invention can also advantageously contain various other, chemically-different salts, of an organic and/or inorganic nature, incorporated to adjust and stabilise the pH value of the solution prepared from the composition. Examples of such salts, which can be chosen both in nature and amount in accordance with the normal skill of those formulating disinfectant compositions therefrom, include alkali metal and alkaline earth metal phosphates, pyrophosphates, polyphosphates, tribicarbonates, polyphosphates, carbonates, 30 borates, acetates, citrates, lactates and tartrates. The proportion in which such inorganic and/or organic salts can be present in the compositions of the invention is again not highly critical, but normally they will be present in an amount within the range of from 1% to 90% (by weight of the whole composition), and preferably in an amount within the range of from 5% to 80% (by weight of the whole composition).

The compositions according to this invention may also often and advantageously incorporate surface-active agents, which serve to enhance the wetting and cleaning action of the aqueous solution prepared from the com-

position.

Suitable anionically-active agents include for instance alkyl sulphates, e.g. coconut oil alkyl sulphates and tallow oil alkyl sulphates; alkyl sulphonates, e.g. lauryl sulphonates; alkylaryl sulphonates, e.g. alkylbenzene sulphonates whose alkyl radicals contain 8 to 14 C atoms; soaps derived from natural or synthetic fatty acids; alkylether sulphates; alkylphenolether sulphates; and alkylsulphosuccinates, e.g. lauryl alcohol sulphosuccinate.

Suitable non-ionically-active agents include for instance alkylphenolpolyglycolether(s), e.g. nonylphenolpolyglycolether(s); fatty alcohol polyglycolethers(s); fatty acid polyglycolester(s); polyoxypropyleneglycols (pluronics); aminoxides, e.g. dodecyldimethylaminoxides; and betaines, e.g. carboxysulphate- or sul-

phonate-betaines.

The proportion in which such surface-

active agents may be present is again not highly critical, but normally will be within the range of from 0.5% to 80% by weight, and preferably from 1% to 50% by weight, based upon the whole composition according to the invention.

The compositions of the invention also may, and often advantageously will, contain further substances which increase the stability of the organic peracid formed during hydrolysis, e.g. complex-formers such as ethylenediaminetetracetic acid and/or its alkali metal and/or

tetracetic acid and/or its alkali metal and/or alkaline earth metal salts, nitrilotriacetic acid and its alkali metal and/or alkaline earth metal salts, alkali metal and/or alkaline earth metal salts of metaphosphoric acid, alkali metal and/or alkaline earth metal salts of

polyphosphoric acid, water-soluble salts of high molecular polycarboxylic acids, alkylphosphonic acids, dialkylphosphonic acids, such as methylenediphosphonic acid, polyphosphonic acids, urea, pyridine-2,3-dicarboxylic acid or pyridine-2,6-dicarboxylic acid.

Some of these stabilizing substances also simultaneously serve a further function, namely in some cases that of binding (by the formation of complexes) any heavy-metal ions which otherwise might form coloured complexes with the aromatic hydroxycarboxylic acids; while in other cases they may make the water soft

by binding the alkaline earth metal ions which cause water-hardness.

These various additional substances, whether serving only as stabilizers or also for other purposes, will normally be present in the composition in a proportion within the the range of from 0.5% to 80% by weight (of the whole composition) and preferably within the narrower range of from 2% to 20% by weight (of the whole composition).

The compositions of the invention may also, and sometimes with advantage, include chemically- and biologically-inert carriers, vehicles or fillers, such as for instance alkali metal and/or alkaline earth metal sulphates, chlorides, silicates, carboxymethylcellulose and water-soluble salts of aromatic sulphonic acids such as benzene-, toluene-, xylene- and

cumenesulphonic acid.

Although such filler etc. substances are chemically- and biologically-inert, they may fulfill other purposes. Thus for instance they can also have the function of binding water in order to hinder agglomeration of the composition otherwise liable to be brought about by atmospheric moisture, by liberated water of crystallization or by any other sources of water. Obviously, the proportion of such filler etcetera substances is not critical, but normally they will be present in a proportion of from 0.5% to 80% by weight, and preferably 2% to 50% by weight, of the whole

composition.

The compositions of the invention may also

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and preferably within the range of from 1% to 20% by weight.
Suitable additional anti-microbial sub-

stances are for example carboxylic acids such as benzoic acid, salicylic acid and sorbic acid, as well as phenols such as 2,6-dimethyl-4-

bromophenol and o-phenylphenol.

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The compositions according to this invention may be prepared in the form of powders or granules on the one hand, or of tablets or like shaped solid bodies on the other hand.

It is one of the advantages of the compositions of the invention that they are practically odourless—and indeed so too are the solutions prepared by dissolving them in water or other aqueous systems. These solutions moreover can be regarded as both physiologically and toxicologically harmless, and have no offensive taste.

The invention of course extends to the disinfectant solutions formed by dissolving the compositions herein disclosed in an aqueous medium.

The organic peracid generated in situ when the composition is dissolved possesses an outstanding and very quickly developed antimicrobial activity, which is most important for the practical use of the compositions of the invention as disinfectants. The fact that the organic peracid is continuously generated for some time after the composition has been dissolved is also important, since the organic peracids are not stable for long, but gradually decompose under the catalytic influence of metal ions, enzymes or other catalyticallyactive substances, to form oxygen and the carboxylic acids from which they are derived.

It is however also a matter of special significance for the practical use of the compositions of the invention that the hydroxyaromatic-carboxylic acid, also generated in situ when the composition is dissolved, equally itself possesses anti-microbial activity, but one rather different from that of the organic peracid. Whereas the percarboxylic acid causes a quick killing of germs, the hydroxy-aromaticcarboxylic acid is relatively slow to act, but

well as walls, floors and other surfaces, especially in hospitals and in medical practice generally, as well as in veterinary work, in the household, in industrial and public health fields and generally in the field of sanitation and hygiene.

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In order that the invention may be more easily understood, it will now be further illustrated by the test results set out below.

Preferred but typical compositions of this invention (namely (1) a mixture of 1.8 grams of acetyl-salicylic acid and 3.0 grams of sodium perborate, and (2) a mixture of 2.4 grams of benzoyl-salicylic acid and 3.0 grams of sodium perborate) were respectively each dissolved in soft water to make 200 grams of solution in each case. The bacteriological activities of those solutions were then investigated as follows:-

bacteriological investigation was The carried out in accordance with the "Guidelines for the Examination of Chemical Disinfectants", 3rd Edition, published on behalf of the DGHM (Deutsche Gesellschaft für Hygiene und Mikrobiologie) by Gustav Fischer Verlag, Stuttgart, Germany, as there laid down for suspension, germ-carriers and 100 surfaces.

According to the DGHM Guidelines a disinfecting agent is effective if it kills the pathogenic germs (a) within 30 minutes in the suspension test and (b) within 6 hours 105 in the surface test, at the same concentrations in each case of the active substance. The germ-carriers test is a more severe one, in which absorbtive materials (such as the mineral granules and cambric respectively 110 used below) are soaked in dispersions of the pathogenic germs before treatment with the active substance.

The results obtained with test solutions formed by dilution of Compositions (1) and 115 (2) are set out respectively in Tables 1 and 2 below. In the Tables the concentration of the test solutions is expressed as a percentage of the concentration of the relevant Composition, arbitrarily taking the concentration of 120 the Composition to be 100%. Within each table the test solutions are divided into three groups, each consisting of three solutions; the first and second groups are solutions which 5 are 30 minutes old at the commencement of

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the test, and the third group is solutions which are 18 hours old at that time. Each solution in the second group of each Table is additionally loaded with 20% of protein (serum).

TABLE 1

		hia		Wood varni sh										
		Escherichia	coli	PVC v	4 h	1h	ᄪ		_					
	ses	. S	2	-		-	-			-	\dashv		-	
	Surfaces	lococa	aureus	Wood varnish										
		Staphylococcus aureus		PVC	Х _р	χ h	1 ^h							
	Germ carriers	Cambric	Californic	Mycobac- terium smegmatis	>120	>120	>120							
	Germ	Gramiles.	Oranuics	Bacil- lus sub- tilis	1h	$_{1}$ h	$^{1}\mathrm{h}$						·	
		gi		Asper- gillus niger	>30	>30	>30							25
		Suspension, fungi		Candida albicans	15	2 1/2	2 1/2							10
88	ŀ	Susp	Tricho-	phyton menta- grophytes	5	2 1/2	2 1/2							5
acid ke up to 200 g				Proteus vulgaris	15	15	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	5
Alsalicylic Arborate H,O to ma			, bacteria	Pyo- genes	2 1/2	2 1/2	2 1/2	2 1/2	2/1/2	2 1/2	2 1/2	2 1/2	2 1/2	10
1.8 g Acetylsalicylic 3.0 g Na-perborate Deioni sed H.O to mah			Suspension, bacteria	Klebsi- ella pneumon.	5	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	5
			, L	Staphylo- coccus aureus	30	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	S	5	2 1/2	5
Composition (1)			% age	ration of test solutions	5	01	25	25	20	80	10	25	20	MIC *

* Minimum inhibitory concentration.

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TABLE 2

2.4 g Benzoylsalicylic acid 3.0 g Na-perborate Deionised H₂O to make up to 200 g Composition (2)

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	Escherichia coli		110	Wood varnish											
	Surfaces	2007	Esch		PVC	4 %	q9	1 p					T		
		Staphylococcus		aureus	Wood vamish										
		Staphy		ä	PVC	ч х	4%	4 %							
	Germ carriers	Combrid	Cambric		terium smegmatis	>120	>120	15							
	Germ	Granning	Granules		lus sub-	ęh	3h	14							
	Suspension, fungi		Acres		gillus niger	>30	30	21/2							25
					Candida albicans	5	2 1/2	2 1/2							25
	Suspensi		Tricho	phyton	menta- grophytes	2 1/2	2 1/2	2 1/2							5
	Suspension, bacteria				Proteus vulgaris	>30	5	5	2 1/2	2 1/2	2 1/2	5	2 1/2	2 1/2	5
					Pyo- genes	>30	2/12	2 1/2	2 1/2	2 1/2	2 1/2	5	2 1/2	2 1/2	S
						5	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	5
				Staphylo- coccus aureus		2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	2 1/2	>30	2 1/2	2 1/2	5
		% age	╶┸┊┈┈╸╎┈╟╍╏╸╏┈┟╼┟╼╏╌╏╺╏╸							*MIC*					

* Minimum inhibitory concentration.

From the results set out in Tables 1 and 2 above, it will readily be seen (a) that 3 Compositions (1) and (2), when dissolved 1 in water, both display a broad spectrum of anti-microbial activity, coupled moreover with an anti-fungal activity; (b) that as can be seen from the results with the 18-hour Ś

2 solutions, the duration of the activity of the solutions is excellent, bearing in mind that to be practically-useful a disinfectant should have a duration of activity of about 1 day or more; and (c) that the solutions have an outstanding ability to withstand loading with protein (serum).

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In this Specification the term "alkyl" represents alkyl groups having from 1 to 18 carbon atoms, preferably from 1 to 12 carbon atoms; 5 the term "lower alkyl", "lower alkoxy" and "lower acyloxy" respectively represent alkyl, alkoxy and acyloxy groups having from 1 to 6 carbon atoms, usually only 1 to 5 carbon atoms, and preferably no more than 4 carbon 10 atoms; and the term lower oxa-alkyl represents a lower alkyl group having a straight chain of at least three carbon atoms in which a carbon atom included in the chain is replaced by an oxygen atom.

The preferred lower alkyl groups are methyl, ethyl and propyl.

WHAT WE CLAIM IS:—

1. Storable compositions, for dilution with water to form anti-microbial solutions, which 20 comprise:—

(A) at least one acyloxy-aromatic-carboxylic acid of the general formula:-

[in which the carboxy group is in the ortho-, 25 meta- or para-position relative to the acyloxy radical; and either R represents a lower alkyl or lower oxa-alkyl radical, optionally substituted by a hydroxy group, a lower alkoxy group, a lower acyloxy group, a carboxy group 30 or a halogen atom, or R represents a phenyl group, optionally substituted by one or more hydroxy group(s), halogen atom(s), lower alkyl group(s) and/or lower alkoxy group(s)] and/or at least one water-soluble alkali metal, 35 alkaline earth metal or ammonium or alkylsubstituted (including quaternary) ammonium salt thereof; and

(B) one or more H₂O₂ source(s). 2. Compositions as claimed in claim 1, 40 in which the radical R in the acyloxy-aromaticcarboxylic acid of general formula I is a methyl, ethyl, hydroxy-methyl, acetoxymethyl, methoxy-methyl, ethoxy-methyl, 1hydroxy-ethyl, 2-hydroxy-ethyl, 2-carboxy-45 ethyl, 3-carboxy-propyl, 3-carboxy-2-oxapropyl, chloro-methyl, phenyl, 2-methyl-phenyl, 3-methyl-phenyl, 4-methyl-phenyl, 2-tert-butyl-phenyl, 3-tert-butyl-phenyl, 4tert-butyl-phenyl, 2-methoxy-phenyl, 3-methoxy-phenyl, 4-methoxy-phenyl, 2-ethoxyphenyl, 3-ethoxy-phenyl, 4-ethoxy-phenyl, 2-chloro-phenyl, 3-chloro-phenyl, 4-chlorophenyl, 2-hydroxy-phenyl, 3-hydroxy-phenyl or 4-hydroxy-phenyl group.

3. Compositions as claimed in claim 1 or claim 2 in which the acyloxy-aromatic-carboxylic acid of general formula I is acetyl-, propionyl-, benzoyl-, (4-methoxy-benzoyl)-, (3-chloro-benzoyl), (4-tert-butyl-benzoyl)- or (4-methyl-benzoyl)-salicyic acid; 3-acetoxy-, 3-propionyloxy-, 3-benzoyloxy-, 3-(4-methoxy-benzoyloxy)-, 3-(3-chloro-benzoyloxy)-, 3-(4-tert-butyl-benzoyloxy)- or 3-(4methyl-benzoyloxy)-benzoic acid; or 4-acetoxy-, 4-propionyloxy-, 4-benzoyloxy-, 4-(4-methoxy-benzoyloxy)-, 4-(3-chloro-benzoyloxy)-, 4-(4-tert-butyl-benzoyloxy) or 4-(4methyl-benzoyloxy)-benzoic acid.

4. Compositions as claimed in any of claims 1 to 3, in which the acyloxy-aromaticcarboxylic acid of general formula I is acetyl-, benzoyl-, (4-methoxy-benzoyl)- or (4-tert-butyl-benzoyl)-salicylic acid; 3-acetoxy-, 3-benzoyloxy- or 3-(4-methoxybenzoyloxy)-benzoic acid; or 4-acetoxy-, 4-propionyloxy-, 4-benzoyloxy- or 4-(4-methoxybenzoyloxy)-benzoic acid.

5. Compositions as claimed in any of claims 1 to 4, in which the acyloxy-aromaticcarboxylic acid of general formula I used as component (A) is present in the form of one of its water-soluble lithium, sodium, potassium, magnesium, ammonium, substituted ammonium or quaternary ammonium salts.

6. Composition as claimed in claim 5, in which the substituted (including quaternary) ammonium salts present are those in which some or all of the hydrogen atoms have been replaced by alkyl radicals.

7. Compositions as claimed in any of claims 1 to 6, in which the acyloxy-aromatic-carboxylic acid salt(s) present is/are the sodium and/or potassium and/or ammonium and substituted (including quaternary) ammonium

8. Compositions as claimed in any of the preceding claims, in which the H2O2 source is or includes one or more alkali metal and/or ammonium perborates, percarbonates, perphosphates, peroxides and/or percarbamides, and/or alkali metal and/or ammonium salts of persulphuric acid and/or peroxydisulphuric acid.

9. Compositions as claimed in any of the preceding claims, in which the acyloxyaromatic-carboxylic acid(s) and/or salts thereof serving as component (A) and the H₂O₂ source(s) serving as component (B) are present in a molar weight ratio within the range of from 1:10 to 10:1.

10. Compositions as claimed in claim 9, in which the molar weight ratio is within the range of from 1:4 to 4:1.

11. Compositions as claimed in any of the preceding claims, which also contain one or 115 more other, chemically-different organic and/ or inorganic salts, chosen in nature and amount to adjust and stabilise the pH value

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of a solution prepared by dissolving the composition in water to a pH value within the range of from 4 to 12. 12. Compositions as claimed in claim 11, in which the other salt(s) include alkali metal and/or alkaline earth metal phosphates, pyrophosphates, polyphosphates, tripolyphosphates, carbonates, bicarbonates, borates, acetates, citrates, lactates and/or tartrates. 13. Compositions as claimed in claim 11 or claim 12, in which the other salt(s) are present in an amount within the range of from 1% to 90% (by weight of the whole composition). 15 14. Compositions as claimed in claim 13, in which the other salt(s) are present in an amount within the range of from 5% to 80% (by weight of the whole composition). 15. Compositions as claimed in any of the preceding claims, which also incorporate surface-active agents, to enhance the wetting and cleaning action of any aqueous solution prepared from the composition. 16. Compositions as claimed in claim 15, which contain anionically-active surface-active agents including one or more alkyl sulphates, alkyl sulphonates, alkylaryl sulphonates, soaps derived from natural or synthetic fatty acids, alkylether sulphates, alkylphenolether sulphates and/or alkylsulphosuccinates. 17. Compositions as claimed in claim 15 or claim 16, which contain non-ionicallyactive surface-active agents including one or alkylphenolpolyglycolether(s), fatty alcohol polyglycolether(s), fatty acid polyglycolester(s), polyoxypropyleneglycol(s), aminoxide(s) and/or betaine(s). 18. Compositions as claimed in any of claims 15 to 17, in which the surface-active agent(s) are present in an amount within the range of from 0.5% to 80% (by weight of the whole composition). 19. Compositions as claimed in claim 18, in which the surface-active agent(s) are present within the range of from 1% to 50% (by weight of the whole composition). 20. Compositions as claimed in any of the preceding claims, which also contain one or more stabilizing substances which increase 50 the stability of the organic peracid formed during hydrolysis. 21. Compositions as claimed in claim 20, in which the stabilizing substance(s) include

ethylenediaminetetracetic acid and/or its alkali metal and/or alkaline earth metal salts, nitrolo-

triacetic acid and/or its alkali metal and/or

alkaline earth metal salts, alkali metal and/or alkaline earth metal salts of metaphosphoric

acid, alkali metal and/or alkaline earth metal

salts of polyphosphoric acid, water-soluble

salts of high molecular weight polycarboxylic

acids, alkylphosphonic acids, dialkylphos-

phonic acids, polyphosphonic acids, urea,

pyridine-2,3-dicarboxylic acid and/or pyridine-2,6-dicarboxylic acid. 65 22. Compositions as claimed in claim 20 or claim 21, in which the stabilizing substance(s) are present in a proportion within the range of from 0.5% to 80% by weight (of the whole composition). 23. Compositions as claimed in claim 22, in which the stabilizing substance(s) are present within the range of from 2% to 20% by weight (of the whole composition). 24. Compositions as claimed in any of the preceding claims which also include chemically- and biologically-inert carriers, vehicles or fillers. 25. Compositions as claimed in claim 24, in which the fillers are so chosen as to hinder agglomeration of the composition by atmospheric or other moisture. 26. Compositions as claimed in claim 24 or claim 25, in which the fillers are present in a proportion of from 0.5% to 80% (by weight of the whole composition). 27. Compositions as claimed in claim 26, in which the fillers are present in a proportion of from 2% to 50%. 28. Compositions as claimed in any of the preceding claims, which also contain corrosion inhibitors and/or perfumes and/or other anti-microbial substances. 29. Compositions as claimed in claim 28, in which the corrosion inhibitor(s) present are benzotriazole, alkali phosphate, alkali hexametaphosphate, alkali nitrate, alkyl phosphate, aminoxide, ammonia soap, sodium silicate, sodium benzoate, sodium fluoride and/or alkylsulphamidocarboxylic acid. 30. Compositions as claimed in claim 29, in which the proportion of corrosion inhibitor(s) present is in the range of from 0.5% to 30% by weight of the whole composition. 31. Compositions as claimed in claim 30, 105 in which the proportion of corrosion inhibitor(s) is in the range of from 1% to 20%. 32. Compositions as claimed in any of claims 28 to 31, in which the other antimicrobial substances present are benzoic acid, 110 salicylic acid, sorbic acid, 2,6-dimethyl-4-

bromophenol and/or o-phenylphenol.

33. Disinfectant solutions formed by dissolving the compositions claimed in any of the previous claims in an aqueous medium.

34. Solutions as claimed in claim 33, having a pH in the range of from 8 to 12. 35. Solutions as claimed in claim 33, having a pH in the range of from 4 to 8.

36. A process for combatting microbial 120 and/or fungal infection in a locus, which comprises the steps of dissolving a composition as claimed in any of claims 1 to 32 in an aqueous medium to form a disinfectant solution, and then applying said disinfectant 125 solution to the locus.

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